

that enriches the isotope-enriched material drawn from at least one of said distillation columns,

wherein said isotope scrambler is connected by a return conduit that returns the enriched material to at least one of said distillation columns.--

REMARKS

In the Office action mailed May 8, 2002, the drawings are objected to because of informalities, the disclosure is objected to because of informalities, claims 12 and 19 are objected to because of informalities, claims 12-17 and 19-24 are rejected under 35 U.S.C. § 112, second paragraph, as being indefinite, claims 12 and 13 are rejected under 35 U.S.C. § 102(b) as being anticipated by Atkinson et al (U.S. Patent No. 4,759,786), claims 12-17 and 19-24 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Shimuzu (U.S. Patent No. 4,173,620) in view of Atkinson, and claims 12-17 and 19-24 are rejected under the judicially created doctrine of obviousness-type double patenting.

In response, applicants have amended the drawings and specifications to correct informalities, have canceled claims 12-17 and 19-24, and have added new claims 37-44.

Regarding Claim Rejection – 35 U.S.C. § 112

Please note that “n” is defined as “n can be set optionally” (ref. : page 27, line 17) in Claim 37.

Please note that “middle section” is indicated in page 21 (line 24) to page 22 (line 3), broken lines in Figure 1, page 24 (lines 18 to 23), and broken lines in Figure 7.

Regarding Claim Rejection – 35 U.S.C. § 102

An apparatus in Claim 37 comprises the return conduits which are connected to an outlet condenser of the $(k+1)^{\text{th}}$ column.

Therefore, it is possible to return a part of the condensed liquid to the upstream column.

In contrast, the return conduits 34, 4, and 6 in the apparatus shown in Figure 5 of U.S. Patent No. 4,759,786 (Atkinson et al) connect the top of column 20 and the bottom of column 8.

As shown in the statement in column 5, lines 48 to 50, the return conduits do not connect condenser 26 and reboiler 44.

Therefore, the citation does not disclose nor suggest the return conduits which connected to an outlet of condenser.

Regarding Claim Rejection – 35 U.S.C. § 103

As mentioned above, an apparatus in Claim 37 comprises the return conduits which are connected to an outlet of condenser of the $(k+1)^{\text{th}}$ column.

On the basis of the difference between the present invention and the citation, it is possible to return a part of the condensed liquid to the upstream column, and it is not necessary to set the pressure in the downstream column higher than the pressure in the upstream column.

Therefore, it is possible to set the pressure within downstream columns lower than in the past, and it is possible to increase the relative vapor pressure between each of the isotopes within each column and to thereby improve the efficiency of the distillation.

As a result, it is possible to reduce both the packing height of each column and the amount of liquid hold-up, and thereby it is possible to shorten the start-up time.

In addition, for the above-mentioned reasons, it is possible to obtain with good efficiency a product containing a high concentration of heavy oxygen isotopes.

In addition, in the return conduits, only the portions corresponding to the liquid head portions which correspond to the difference in pressure of these columns are full of liquid.

In other words, of the return conduits from a downstream column to an upstream column, only the portions of conduits corresponding to liquid head portions which correspond to the pressure difference of those columns for which the return of the return liquid liquefied in the condensers to the upstream column is necessary are filled with liquid.

In addition, in conventional apparatuses (Figure 19), all of the supply conduits from an upstream column to a downstream column are filled with liquid.

In contrast, in the apparatus of the present invention, introduction conduits are filled with vapor, because they are connected to an outlet of the reboiler of the k^{th} column.

For this reason, the amount of liquid hold-up is reduced, and it is possible for the start-up time for the apparatus to be shortened even more.

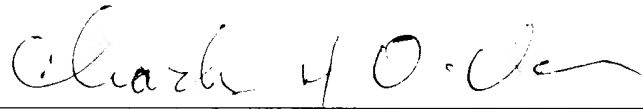
As required by 37 C.F.R. § 1.121, applicants have provided a separate marked-up version of the amended claims showing the changes relative to the previous version of those claims (attached).

The above amendments and remarks are believed to address fully the Examiner's rejections, and place the application in condition for allowance. A prompt indication of the same respectfully is requested. The Examiner is encouraged to telephone

the undersigned if any issues remain that may be resolved by a telephonic interview.

Respectfully submitted,

KOLISCH HARTWELL, P.C.



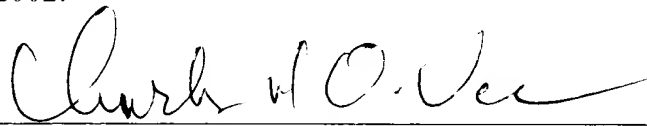
Charles H. DeVoe
Registration No. 37,305
Attorney for Applicants
Customer No. 23581
520 S.W. Yamhill Street, Suite 200
Portland, Oregon 97204
Telephone: (503) 224-6655
Facsimile: (503) 295-6679



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Charles H. DeVoe
Date of Signature: November 5, 2002

VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the Specification:

On page 1, please amend the paragraph beginning on line 21, as follows.

(Amended) In this case, generally, packed columns are used in order to suppress pressure loss within the column.[,] [h]However, it is not uncommon for the required packing height therefor to reach several hundred meters.

On page 2, please amend the paragraph beginning on line 15, as follows.

(Amended) In addition, in general, as in isotope separation processes, when the relative vapor pressure of the separation components is extremely small and the packing height is extremely large, the time (hereinafter referred to as the start-up time) from the start up of the apparatus until it becomes possible to collect the stipulated amount (the product amount extracted in accordance with the specifications, or the planned value) may take from several months to several years. Consequently, reduction of the start-up time has hitherto been an issue.

On page 2, please amend the paragraph beginning on line 23, as follows.

(Amended) Figure 19 shows a distillation process having~~(a)~~ the same functions as the apparatus shown in Figure 18, and each column 71, 72, and 73 is~~[are]~~ provided with condensers 5, 7 and 9, and reboilers 6, 8, and 10. These are apparatuses which are generally used in processes comprising a plurality of distillation columns. The diameter of the distillation columns becomes smaller from the column 71, to which the starting material is fed, to the column 73, which is downstream from column 71.

On page 3, please amend the paragraph beginning on line 1, as follows.

(Amended) However, even when using this type of apparatus, since the pressure within the distillation column increases from the first column 71 to the third column 73, in the same way as with the apparatus shown in Figure 18, the distillation efficiency is lower in the second column 72 than in the first column 71, and is lower in the third column 73 than in the second column 72.

On page 3, please amend the paragraph beginning on line 5, as follows.

(Amended) Reductions in the distillation efficiency ~~[are led]~~lead to increases in the necessary packing height of the distillation column and to increases in the process liquid hold up~~[,]~~ ~~[t]~~Therefore, they are not desirable from the point of view ~~[or]~~of shortening the start-up time.

On page 3, please amend the paragraph beginning on line 8, as follows.

(Amended) In addition, in conventional isotope distillation processes, columns packed using unstructured packing have been used. In general, unstructured packing has a larger specific surface area compared with structured packing[.]. [h]However, the liquid hold-up within the distillation column is 10 to 20% of the volume of the column, and in some cases exceeds 20%, and this is a cause of prolonged start-up time.

On pages 4-5, please amend the paragraph beginning on line 22, as follows.

(Amended) The method of enrichment of oxygen isotopes of the present invention is one in which an oxygen-[]starting material containing heavy oxygen isotopes is enriched by means of a cascade process using a plurality of distillation columns (a first column to an n^{th} column) comprising supplying at least a part of the vapor from the bottom of a k^{th} ($1 \leq k \leq (n-1)$) column or an outlet of a reboiler provided in the vicinity of the bottom of the k^{th} column to the top of a $(k+1)^{\text{th}}$ column, an inlet of a condenser provided in the vicinity of the top of the $(k+1)^{\text{th}}$ column, or a middle section of the $(k+1)^{\text{th}}$ column; returning at least a part of the liquid from the top of the $(k+1)^{\text{th}}$ column or an outlet of the condenser of the $(k+1)^{\text{th}}$ column to an inlet of a reboiler of the k^{th} column, the bottom of the k^{th} column, or the middle section of the k^{th} column; and thereby carrying out enrichment in at least one type of oxygen molecule of $^{16}\text{O}^{17}\text{O}$, $^{16}\text{O}^{18}\text{O}$, $^{17}\text{O}^{17}\text{O}$, $^{17}\text{O}^{18}\text{O}$, and $^{18}\text{O}^{18}\text{O}$, which contain heavy oxygen isotopes.

On page 5, please amend the paragraph beginning on line 4, as follows.

(Amended) A method of enrichment of oxygen isotopes in which an oxygen-[]starting material containing heavy oxygen isotopes is enriched by means of a cascade process using a plurality of distillation columns (a first column to an n^{th} column) comprising supplying at least a part of the vapor from the bottom of a k^{th} ($1 \leq k \leq (n-1)$) column or an outlet of a reboiler provided in the vicinity of the bottom of the k^{th} column to the top of a $(k+1)^{\text{th}}$ column, an inlet of a condenser provided in the vicinity of the top of the $(k+1)^{\text{th}}$ column, or a middle section of the $(k+1)^{\text{th}}$ column, pressurizing at least a part of a vapor drawn off from the top of a $(k+1)^{\text{th}}$ column or a vapor from an inlet of the condenser of the $(k+1)^{\text{th}}$ column by means of a blower, and then returning a said vapor to the bottom of the k^{th} column or the middle section of the k^{th} column, and thereby carrying out enrichment in at least one type of oxygen molecule of $^{16}\text{O}^{17}\text{O}$, $^{16}\text{O}^{18}\text{O}$, $^{17}\text{O}^{17}\text{O}$, $^{17}\text{O}^{18}\text{O}$, and $^{18}\text{O}^{18}\text{O}$, which contain heavy oxygen isotopes.

On page 5, please amend the paragraph beginning on line 15, as follows.

(Amended) In addition, the method of the present invention is a method for enrichment in isotopes of oxygen comprising subjecting an oxygen isotope-[]enriched material enriched by means of the above mentioned enrichment method to oxygen isotope scrambling to obtain an enriched product having an even higher concentration of at least one type of the above[-]mentioned oxygen molecules which contain heavy oxygen isotopes.

On page 5, please amend the paragraph beginning on line 20, as follows.

(Amended) In addition, the method of the present invention is a method for enrichment in isotopes of oxygen comprising subjecting an oxygen isotope-[]enriched material enriched by means of the above_[-]mentioned method of enrichment to oxygen isotope scrambling, to obtain an enriched material having a higher concentration of at least one type of said oxygen molecules which contain heavy oxygen isotopes; and obtaining an enriched product having an even higher concentration of at least one type of the above _[-]mentioned oxygen molecules which contain heavy oxygen isotopes by means of conducting the above_[-]mentioned method of enrichment again on said enriched material.

On pages 5-6, please amend the paragraph beginning on line 27, as follows.

(Amended) In addition, the method of producing heavy oxygen water of the present invention comprises obtaining an enriched material which has been enriched in at least one component from oxygen molecules which contain heavy oxygen isotopes by means of cryogenic distillation of an oxygen-[]starting material which contains heavy oxygen isotopes using the above_[-]mentioned apparatus; obtaining water containing heavy oxygen water corresponding to said enriched material by adding hydrogen to said enriched material. Thereafter, this heavy oxygen water is further enriched using the above_[-]mentioned apparatus.

On page 6, please amend the paragraph beginning on line 6, as follows.

(Amended) In addition, the method of the present invention is a method of producing heavy oxygen water in which a water-[]starting material containing heavy oxygen water is enriched by means of a cascade process using a plurality of distillation columns comprising supplying at least a part of the water vapor from the bottom of a k^{th} ($1 \leq k \leq (n-1)$) column, or an outlet of a reboiler provided in the vicinity of the bottom of the k^{th} column to the top of a $(k+1)^{\text{th}}$ column, an inlet of a condenser provided in the vicinity of the top of the $(k+1)^{\text{th}}$ column, or a middle section of the $(k+1)^{\text{th}}$ column, introducing at least a part of the water from the top of the $(k+1)^{\text{th}}$ column, or an outlet of the condenser of the $(k+1)^{\text{th}}$ column into an inlet of a reboiler of the k^{th} column, the bottom of the k^{th} column, or the middle section of the k^{th} column, and thereby carrying out enrichment in at least one type of heavy oxygen water of H_2^{17}O , H_2^{18}O , D_2^{17}O , D_2^{18}O , DH^{17}O , DH^{18}O , which contain heavy oxygen isotopes.

On page 6, please amend the paragraph beginning on line 17, as follows.

(Amended) In the present invention, [since an introduction conduit which connects the bottom of the k^{th} column ($1 \leq k \leq (n-1)$) or an outlet of a reboiler provided in the vicinity of the k^{th} column to the top of the $(k+1)^{\text{th}}$ column, or the middle section of the $(k+1)^{\text{th}}$ column, and a return conduit which connects the outlet of the condenser of the $(k+1)^{\text{th}}$ column to the inlet of a reboiler provided in the vicinity of the bottom of the k^{th}

column, the bottom of the k^{th} column, or the middle section of the k^{th} column are provided,] it is possible to return a part of the liquid drawn off from the condenser of the $(k+1)^{\text{th}}$ column via a[this] return conduit. More particularly, an introduction conduit may connect the bottom of the k^{th} column ($1 \leq k \leq (n-1)$) or an outlet of a reboiler provided in the vicinity of the k^{th} column to the top of the $(k+1)^{\text{th}}$ column. Alternatively, the introduction conduit may connect to the middle section of the $(k+1)^{\text{th}}$ column. Similarly, a return conduit may connect the outlet of the condenser of the $(k+1)^{\text{th}}$ column to the inlet of a reboiler provided in the vicinity of the bottom of the k^{th} column, the bottom of the k^{th} column, or the middle section of the k^{th} column.

On page 8, please amend the paragraph beginning on line 22, as follows.

(Amended) Figure 18 is a schematic structural diagram showing a prior art example of a conventional apparatus.

On page 8, please amend the paragraph beginning on line 24, as follows.

(Amended) Figure 19 is another schematic structural diagram showing another prior art example of a conventional apparatus.

On page 9, please amend the paragraph beginning on line 1, as follows.

(Amended) Figure 1 shows a first embodiment of the apparatus of the present invention. The apparatus shown here is equipped with three distillation columns, a first column 1, a second column 2, and a third column 3, constructed in a cascade[d].

On page 9, please amend the paragraph beginning on line 4, as follows.

(Amended) Here, “constructed in a cascade” means that the distillation columns are connected in such a way that the enriched product of one column can be further enriched in the next distillation column, and then can be further enriched again in the next distillation column, and all the distillation columns together constructed in this way are referred to as a cascade process.

On page 9, please amend the paragraph beginning on line 8, as follows.

(Amended) In addition, in the figure, the path indicated by the broken line is a modification of the present embodiment, and is not included in the apparatus of the present embodiment.

On page 10, please amend the paragraph beginning on line 3, as follows.

(Amended) As reboilers 6, 8, and 10, it is preferable to use a plate fin type reboiler[s]. In addition, the position of installation of these reboilers is such that the amount

of liquid which accumulates at the bottom of each column is reduced to a minimum range within which operation is possible in order to reduce the liquid hold up.

On page 10, please amend the paragraph beginning on line 10, as follows.

(Amended) In the distillation column of the present invention, the outlet (the outlet of first conduit 6a) of reboiler 6 of the first column₁ which is provided in the vicinity of the bottom of the first column 1, and the inlet of the first conduit 7a of the condenser 7 of the second column₂ which is provided in the vicinity of the top of the second column₂ are connected by a first introduction conduit 12 via a valve 12v.

On page 10, please amend the paragraph beginning on line 18, as follows.

(Amended) In addition, the outlet (the outlet of first conduit 7a) of the condenser 7 of the second column and the inlet (the inlet of the first conduit 6a) of the reboiler 6 of the second column are connected by means of the [second]first return conduit 14 via valve 14v.

On page 11, please amend the paragraph beginning on line 3, as follows.

(Amended) [As this structured packing 11, n]Non-promoting-fluid-dispersion type structured packing and/or promoting-fluid-dispersion type structured

packing can be [suitable]suitably used as structured packing 11.

On page 11, please amend the paragraph beginning on line 22, as follows.

(Amended) In addition, the non-promoting-fluid-dispersion type structured packing 52 shown in Figure 3 is a lattice structure comprising of plurality of mutually parallel plates 52a and a plurality of plates 52b which are arranged at right angles with respect to the plates 52a, and this lattice structure is positioned along the direction of the column axis.

On page 11, please amend the paragraph beginning on line 26, as follows.

(Amended) Promoting-fluid-dispersion type structured packing has a shape and structure with which vapor-liquid contact occurs mainly on the surface of the above [-]mentioned structured packing between the liquid descending within the distillation column and the vapor ascending within the distillation column, at which time[,], the liquid and the vapor flow in opposition to one another on the surface of the above [-]mentioned structured packing in the direction of the main flow, which is along the direction of the column axis, and, at the same time, mixing of the liquid and/or the vapor in a direction at right angles to the above [-]mentioned main flow direction is promoted and vapor-liquid contact occurs. These are called structured packing or regular packing in which thin plates of aluminum, copper, aluminum-copper alloy, stainless steel, various plastics, or the like are formed into a variety

of regular forms, and then made into a laminated block.

On page 12, please amend the paragraph beginning on line 10, as follows.

(Amended) In the promoting-fluid-dispersion type structured packing 53 shown in Figure 4, a plurality of wave-shaped thin plates are disposed parallel to the column axis and made into the form of a block by layering the plates so that they come into contact with one another. The wave-shaped grooves in each of the thin plates are inclined with respect to the column axis, and neighboring wave-shaped thin plates are disposed so that the direction of their wave-shaped grooves intersects one another. In addition, a plurality of holes 53a are provided in the thin plates. When the thin plates are disposed perpendicular [with respect] to the horizontal plane, the holes are provided with an interval of spacing there[] between and form a plurality of rows along a direction which is at right angles to the column axis.

On page 12, please amend the paragraph beginning on line 19, as follows.

(Amended) Figure 5 shows an example of a structural unit of another promoting-fluid-dispersion type structured packing. In the promoting-fluid-dispersion type structured packing 54 shown here, a thin plate is molded by being pressed into a wave shape to form wave-shaped grooves. In addition, this example has the feature that extremely small wave-shaped grooves 54b are formed in the thin plates at a fixed angle with respect to the

wave-shaped grooves. In addition, reference 54a indicates holes formed in the thin plate.

On page 13, please amend the paragraph beginning on line 8, as follows.

(Amended) The use of oxygen with high purity is preferred as the above [-]mentioned oxygen vapor starting material. As the above [-]mentioned oxygen of high purity, it is possible to use oxygen with a purity of 99.999% or higher, from which impurities such as argon, hydrocarbons, krypton, xenon, and fluorine compounds (such as perfluorocarbons) have been removed in advance. In particular, the use of an oxygen-[-]starting material from which hydrocarbons have been removed is preferable from the point of view of safety.

On page 13, please amend the paragraph beginning on line 14, as follows.

(Amended) The oxygen-[-]starting material vapor supplied to the first column 1 is distilled by means of vapor-liquid contact with a reflux liquid (i.e., descending liquid) described below, when ascending within the first column 1 and passing through the packing 11.

On page 13, please amend the paragraph beginning on line 17, as follows.

(Amended) The oxygen molecules in the oxygen-[-]starting material vapor

which contain heavy oxygen isotopes (i.e., $^{16}\text{O}^{17}\text{O}$, $^{16}\text{O}^{18}\text{O}$, $^{17}\text{O}^{17}\text{O}$, $^{17}\text{O}^{18}\text{O}$, and $^{18}\text{O}^{18}\text{O}$) are more likely to condense due to their high boiling points, and in the process of vapor-liquid contact, the concentration of heavy oxygen isotopes in the descending liquid increases, and the concentration of heavy oxygen isotopes in the ascending vapor decreases.

On page 13, please amend the paragraph beginning on line 22, as follows.

(Amended) The ascending vapor with reaches the top of the column and which has a reduced concentration of heavy isotopes is drawn off from the column 1 via conduit 21, and is divided into two parts. One part is introduced into the first conduit 5a of the first column condenser 5 where it [is] exchanges heat with the medium for heat exchange flowing within the second conduit 5b, condenses, and is returned to the top of column 1 as reflux liquid. The other part is discharged out of the system via conduit 22 as exhaust vapor 103.

On pages 13-14, please amend the paragraph beginning on line 27, as follows.

(Amended) The reflux liquid returned to the top of the first column 1 flows down as descending liquid over the structure packing 11 while making vapor-liquid contact with the oxygen-[]starting material vapor and the below_[-]mentioned ascending vapor which are ascending within the column 1, and reaches the bottom of the column 1.

On page 14, please amend the paragraph beginning on line 6, as follows.

(Amended) Liquid which reaches the bottom of the first column 1 (hereinafter, the liquid which accumulates at the bottom of each of the columns is referred to as the “column bottom liquid”) is drawn off from the column 1 through conduit 23, and is combined with the below_[-]mentioned first return liquid 107 in the above_[-]mentioned [first]second return conduit 14, and then introduced into the first conduit 6a of the first column reboiler 6, where it is vaporized by heat exchange with the medium for heat exchange flowing within the second conduit 6b, and then drawn off from the reboiler 6 via the above_[-]mentioned first introduction conduit 12 as first column output vapor 102 which is an enriched material enriched in heavy oxygen isotopes.

On page 15, please amend the paragraph beginning on line 5, as follows.

(Amended) The other part of the condensed liquid which was divided after being drawn off from the condenser 7 is supplied to the top of the second column 2 as second column reflux liquid or condensed second column feed vapor 104 through conduit 25, and flows down as descending liquid while making vapor-liquid contact with the ascending vapor within the second column 2 to reach the bottom of the second column 2.

On page 17, please amend the paragraph beginning on line 12, as follows.

(Amended) In more detail, if ☐the flow rate of the desired component accompanying the product 110 collected on☐the outside of the apparatus from the second column or third column☐ divided by ☐the flow rate of the desired component accompany the second column feed vapor 104☐ is equal to ☐the yield of the desired component☐, it is preferable for the setting to be such that the yield of this desired component will☐ be 1% to 10% or less.

On pages 17-18, please amend the paragraph beginning on line 28, as follows.

(Amended) In more detail, if ☐the flow rate of the desired component accompanying the product 110 collected on☐the outside of the apparatus from the third column☐ divided by ☐the flow rate of the desired component accompany the third column feed vapor 108☐ is equal to ☐a yield of the desired component☐, it is preferable for the setting to be such that the yield of the desired component will☐ be 1% to 10% or less.

On page 18, please amend the paragraph beginning on line 21, as follows.

(Amended) It is difficult to adjust all four simultaneously.[,] [t]Therefore, basically, in each column, 1 and 3 are fixed and 2 and 4 are adjusted while watching the condition of the apparatus.

On page 18, please amend the paragraph beginning on line 25, as follows.

(Amended) It [is maintaining]maintains the amount of heat exchange in the reboiler after it has been set in advance so that the vapor load of each column is at a suitable value (planned value). It is preferable for the setting to be such that the density-[]correlated superficial velocity (hereinafter referred to as the superficial F factor) is $0.5 \text{ m/s(kg/m}^3)^{0.5}$ or greater and $3.0 \text{ m/s(kg/m}^3)^{0.5}$ or less, preferably $0.7 \text{ m/s(kg/m}^3)^{0.5}$ or greater and $2.2 \text{ m/s(kg/m}^3)^{0.5}$ or less.

On page 19, please amend the paragraph beginning on line 10, as follows.

(Amended) Next, (4) the amount of heat exchange in the condenser (the pressure and flow rate of the cold medium) is set. This is set such that the pressure within each of the distillation columns [are]is constant. Since (3) the amount of heat exchange in the reboilers is fixed, if the (4) amount of heat exchange in the condenser is small, the pressure in the distillation columns continues to increase.

On page 19, please amend the paragraph beginning on line 26, as follows.

(Amended) The degree of opening of these valves is adjusted while measuring the liquid level at the bottom of each column[s].

On page 20, please amend the paragraph beginning on line 13, as follows.

(Amended) In the apparatus of the present embodiment, since the outlet of the first column reboiler 6 and the inlet of the second column condenser 7 are connected by the first introduction conduit 12 and the outlet of the second column reboiler 8 and the inlet of the third column condenser 9 are connected by the second introduction conduit 13, and since the outlet of the second column condenser 7 and the inlet of the first column reboiler 6 are connected by [first]second return conduit 14[:]; the outlet of the third column condenser 9 and the inlet of the second reboiler 8 are connected by the second return conduit 15, it is possible to return a part of the condensed liquid which has passed through the condensers 7 and 9 as return liquid 107 and 112 to the reboilers 6 and 8.

On page 20, please amend the paragraph beginning on line 21, as follows.

(Amended) For this reason, it is possible to set the pressure within columns 2 and 3 lower than in the past, and it is possible to increase the relative vapor pressure between each of the isotopes within each column [and]thereby to improve the efficiency of the distillation. As a result, it is possible to reduce both the packing height of each column[, it is possible to reduce]and the amount of liquid hold-up, and thereby it is possible to shorten the start-up time.

On page 21, please amend the paragraph beginning on line 9, as follows.

(Amended) In particular, when the diameter of the distillation columns is

small, the diameter of the piping of the return conduits is [relative]relatively large[.].
[t]Therefore, the difference between the liquid hold-up of the apparatus of the present invention and that of a conventional apparatus is considered to be large. In addition, from the point of view of the start-up time, the superiority of the apparatus of the present invention is considered to be even more obvious.

On pages 21-22, please amend the paragraph beginning on line 24, as follows.

(Amended) In the distillation column of the above__[-]described embodiment, the outlet of the second column condenser 7 and the inlet of the first column reboiler 6 are connected by the first return conduit 14 and the outlet of the third column condenser 9 and the inlet of the second column reboiler 8 are connected by the return conduit 15. However, the apparatus of the present invention is not limited to this embodiment, and the first conduit can be used to connect the outlet of the second condenser 7 and the middle section of the first column 1 as shown by reference 14a (the conduit indicated by the broken line), and the second return conduit can be used to connect the outlet of the third column condenser 9 and the middle section of the second column 2, as shown by reference 15a (the conduit indicated by the broken line).

On page 22, please amend the paragraph beginning on line 6, as follows.

(Amended) Figure 7 shows a second embodiment of the apparatus of the

present invention. The apparatus shown here is different from[to] the apparatus shown in Figure 1 on the following points.

On page 23, please amend the paragraph beginning on line 1, as follows.

(Amended) An oxygen vapor-[]starting material supplied to the inside of the first column 1 as first column feed 101 via a conduit 20 is distilled by means of vapor-liquid contact within the first column 1.

On page 24, please amend the paragraph beginning on line 10, as follows.

(Amended) In the apparatus of the above-described embodiment, the outlet of the first column re boiler 6 and the top of the second column 2 are connected by means of the first introduction conduit 35, and the outlet of the second column reboiler 8 and the top of the third column 3 are connected by means of the second introduction conduit 36, in addition, the outlet of the second column condenser 7 and the bottom of the first column 1 are connected by means of the return conduit 37, and the outlet of the third column condenser 9 and the bottom of the second column 2 are connected by means of the second return conduit 38. However, the apparatus of the present invention is not limited to this embodiment.

On page 25, please amend the paragraph beginning on line 7, as follows.

(Amended) [As blowers 41 and 42, n]Normal temperature compressors or low temperature compressors can be used as blowers 41 and 42. However, when a normal temperature compressor is used, as shown in Figure 9, it is necessary to have heat exchanges 41a and 42a in the return conduits 37' and 38'.

On page 25, please amend the paragraph beginning on line 21, as follows.

(Amended) In the same way, the column top vapor 111 drawn off from the top of the third column 3 is divided after being drawn off from the third column 3. One part is considered in the third column condenser 9, and then return to the top of the third column to become reflux liquid for that column. The other part passes along the second return conduit 38 and reaches the blower 42 when it is pressurized and then returned to the bottom of the second column 2 as second return liquid 112".

On pages 25-26, please amend the paragraph beginning on line 27, as follows.

(Amended) In the apparatus of the present embodiment, since the supply for each conduit is in vapor form, and not a liquid form, it is possible to shorten the start-up time in the same way as for the apparatus of the above[-]mentioned first embodiment. In addition, since the bottoms of the columns are connected to the tops of the columns and the tops of the columns are connected to the bottoms of the column, respectively, it is possible to obtain a product which has a high concentration of heavy oxygen isotopes.

On page 26, please amend the paragraph beginning on line 18, as follows.

(Amended) In the above_[-]described embodiment, an apparatus having three distillation columns is shown.[,] [h]However, in the apparatus of the present invention, the number of distillation columns is not limited to this embodiment.

On page 27, please amend the paragraph beginning on line 6, as follows.

(Amended) This circulation conduit 81 is such that the medium for heat exchange within the storage tank 82 is drawn off, passes through the pump 83 and the first passage of the [super cooler]subcooler 84, and is introduced into the condensers B_1 to B_n . Then, it passes through the second passage of the [super cooler]subcooler 84 and reaches the first conduit of the heat exchanger 85. Next, it passes through the blower 86 and the second passage of the heat exchanger 85, is introduced into the reboilers C_1 to C_n , and then returned to the storage tank 82. In addition, the [super cooler]subcooler 84 is for the purpose of cooling the medium for heat exchange prior to its reaching the condensers.

On page 27, please amend the paragraph beginning on line 18, as follows.

(Amended) In addition, the above_[-]described embodiment shows a method for enrichment in the heavy isotopes of oxygen by means of cryogenic distillation of an oxygen-[-]starting material. However, the present invention is not limited to this embodiment. It

is also possible to produce heavy oxygen water by carrying out distillation using water as the starting material and using the apparatus of the above [-]described embodiments to distill this water to enrich it in at least one type of heavy oxygen water of H_2^{17}O , H_2^{18}O , D_2^{17}O , D_2^{18}O , DH^{17}O and DH^{18}O , which contain heavy oxygen isotopes.

On page 28, please amend the paragraph beginning on line 7, as follows.

(Amended) The hydrogenation reaction device 300A shown in Figure 13 comprises a buffer tank 341 for temporarily storing the enriched material (oxygen[vapor]) which has passed through the oxygen distillation column unit F_1 ; a conduit [342]44 for introducing the oxygen [vapor] into the buffer tank 341; a conduit 343 for introducing hydrogen (or deuterium) supplied from a supply source not shown in the figure; a combustion chamber 344 for reacting the oxygen and hydrogen supplied from the conduits 342 and 343; and a combustor 300 having a controller 345.

On page 28, please amend the paragraph beginning on line 14, as follows.

(Amended) The combustion chamber 344 comprises a burner 344a for mixing and combusting oxygen and hydrogen supplied into the combustion chamber 344; a heater 344b for igniting the oxygen-hydrogen mixed [vapor]gas; and a cooling coil 344c for cooling the reactant product (i.e., water vapor). Additionally, reference 344d indicates a discharge opening which is provided for expelling the reactant product (water) of the

combustion chamber 344 via a valve.

On page 28, please amend the paragraph beginning on line 19, as follows.

(Amended) The controller 345 regulates a flow control valve 342b by means of signals based on the flow rate of oxygen [vapor]measured by an oxygen flow rate detector 342a provided in conduit 342, and thereby the controller 345 is able to adjust the supply flow rate of oxygen [vapor]supplied into the combustion chamber 344 via the conduit 342.

On page 28, please amend the paragraph beginning on line 27, as follows.

(Amended) Further, references 342c and 343c indicate check valves; references 342d and 343d indicate back-fire prevention chambers; and reference 344e indicates a conduit for discharging the small amount of unreacted [vapor]gas remaining in the combustion chamber 344, via a valve.

On page 29, please amend the paragraph beginning on line 9, as follows.

(Amended) The oxygen [vapor]introduced into the combustor 300 passes through the buffer tank 341, and is introduced into the combustion chamber 344 via the conduit 342 and through the burner 344a.

On page 29, please amend the paragraph beginning on line 14, as follows.

(Amended) At this point, the controller 345 performs a calculation based on signal based on a predetermined value and a feedback signal based on the flow rate of oxygen [vapor] measured by the oxygen flow rate detector 342a. The controller regulates the flow rate control valve 342b by means of a signal resulting from this calculation. In the same manner, controller 345 performs a calculation based on the signal based on a predetermined value and a feedback signal based on the flow rate of hydrogen measured by the hydrogen flow rate detector 343a. The controller regulates the flow rate control valve 343b by means of a signal resulting from this calculation. As a result, the aforementioned oxygen and hydrogen are supplied into the combustion chamber 344 in volumes which approximate the stoichiometric volume for producing water.

On page 29, please amend the paragraph beginning on line 21, as follows.

(Amended) The oxygen and hydrogen supplied to the combustion chamber 344 are always regulated to a volume which closely approximates the aforementioned stoichiometric volume by means of the aforementioned feedback control. However, despite this, excess supplied vapor is regularly discharged via a valve from a discharge conduit 344e, and this prevents the gas[vapor] from accumulating in the combustion chamber 344.

On page 29, please amend the paragraph beginning on line 26, as follows.

(Amended) In order to further reduce the volume of the exhaust gas[vapor], it is preferable to jointly employ an even more precise controlling means, such as a feed-forward controlling method. The aforementioned oxygen and hydrogen supplied into the combustion chamber 344 are mixed by means of the burner 344a, and subsequently, jetted into the combustion chamber 344, ignited by means of the heater 344b, and reacted with each other to produce water.

On page 30, please amend the paragraph beginning on line 4, as follows.

(Amended) Most of the produced water is condensed by the cooling coil 344c, and subsequently expelled from[out of] the combustion chamber 344 via the discharge opening 344d. and introduced into the water distillation column unit F₂ through conduit 45 as produced water 113.

On page 30, please amend the paragraph beginning on line 7, as follows.

(Amended) Since this produced water 113 is obtained using the intermediate product vapor 110', which is a enriched material in heavy oxygen isotopes, it is heavy oxygen water which contains a large number[amount] of heavy oxygen isotopes.

On pages 30-31, please amend the paragraph beginning on line 26, as follows.

(Amended) The isotope scrambler 47 is for further enriching the heavy oxygen isotope enriches material enriched by means of the columns $A_l[l]$ to A_h by means of oxygen isotope scrambling. The inlet side of the isotope scrambler is connected to the bottom of the h^{th} column A_h via the extraction conduit 48, and the outlet side of the isotope scrambler is connected to the middle section of the $i^{th}[th]$ column $A_i[i]$ which is at a later stage and adjacent to $h^{th}[th]$ column $A_h[h]$ via a return conduit 49.

On page 31, please amend the paragraph beginning on line 3, as follows.

(Amended) As with the isotope scrambler 47, in addition to the use of an isotope exchange reaction catalyst, it is possible to temporarily form other compounds from the oxygen molecules, and then subsequently break these compounds down to obtain oxygen molecules.

On page 31, please amend the paragraph beginning on line 6, as follows.

(Amended) When using the former of these, as the isotope scrambler 47, the column bottom vapor of column $A_h[h]$ which is an oxygen isotope enriched material is brought into contact with an isotope exchange reaction catalyst, to promote isotope exchange reactions discussed below within the enriched material, and thereby it is possible to further increase the concentration of molecules of heavy oxygen isotopes within the enriched material.

On page 31, please amend the paragraph beginning on line 11, as follows.

(Amended) In this situation, the isotope scrambler 47 is provided with a catalytic column (not shown in the figure), and an isotope exchange reaction catalyst which is packed inside this catalytic column.

On page 31, please amend the paragraph beginning on line 16, as follows.

(Amended) In addition, as the isotope exchange reaction catalysts, in addition to those mentioned above, it is possible to use a catalyst including at least one type selected from the group comprising Ti-oxide, Zr-oxide, Cr-oxide, Mn-oxide, Fe-oxide, Co-oxide, Ni-oxide, Cu-oxide, Al-oxide, Si-oxide, Sn-oxide, and V-oxide. In addition, it is possible to use one of either of the above-mentioned single catalysts or the above-mentioned metal oxide catalysts or to use a mixture of a plurality of them.

On page 31, please amend the paragraph beginning on line 22, as follows.

(Amended) As with the isotope scrambler 47, when the later is being used, the column bottom vapor of column $A_h[h]$ which is a heavy oxygen isotope enriched material is temporarily converted to a different compound (for example, water), this is subsequently broken down to give molecular oxygen, and thereby the concentration of molecules of heavy oxygen isotope molecules within the enriched material is further

increased. An example of the isotope scrambler used in this method is shown in Figure 15.

On page 32, please amend the paragraph beginning on line 9, as follows.

(Amended) The argon circulation blower 87 is provided in order to circulate the gas[vapor] within the argon circulation system having a catalytic column 88, a cooler 91, and a chiller 92. Since oxygen and hydrogen coexist within this circulation system, in consideration of the explosibility range, an argon supply conduit 98 for supplying argon for diluting the vapor within the circulation system is connected to the circulation system.

On page 32, please amend the paragraph beginning on line 14, as follows.

(Amended) In addition, this circulation system is provided with a hydrogen [vapor] supply conduit 97 for the purpose of maintaining the concentration of hydrogen at the outlet of the chiller 92 at approximately 2%.

On page 32, please amend the paragraph beginning on line 20, as follows.

(Amended) Feed 101 is supplied to first column A₁ through conduit 20, and, thereafter, it is enriched in heavy oxygen isotopes by following the above[-]described process until it reaches the column A_h[h], and thereby a column bottom liquid of column A_h[h] from which [is] an enriched material is obtained.

On page 32, please amend the paragraph beginning on line 24, as follows.

(Amended) Next, this column bottom liquid is introduced to the isotope scrambler 47 as introduction oxygen[vapor] 115 through the conduit 48.

On page 33, please amend the paragraph beginning on line 5, as follows.

(Amended) This gaseous[vapor] mixture is cooled by means of the cooler 91 and the chiller 92, and the water is separated into the storage tank 93. This water is sent to the electrolysis tank 94 (pressure 400 kPa gauge) by means of a pump provided within the electrolysis apparatus 89, undergoes electrolysis, and is separated into hydrogen [vapor] containing a small amount of oxygen and oxygen vapor containing a small amount of hydrogen again. The former hydrogen [vapor] is recovered from the above_[-]described argon circulation system. The small amounts of hydrogen and water in the latter oxygen [vapor] are removed by means of an oxygen purifier 90 which has a catalytic column 95 and an adsorber 96, and the oxygen [vapor] is drawn off through conduit 49 and returned to the oxygen distillation column.

On page 33, please amend the paragraph beginning on line 15, as follows.

(Amended) In order to prevent loss of oxygen vapor which is enriched in heavy oxygen isotopes within the regenerated gas[vapor] of the adsorber 96, separate super

high-[]purity oxygen [vapor]([a vapor]having the same specifications as the first column feed vapor of the distillation column) is used.

On page 33, please amend the paragraph beginning on line 19, as follows.

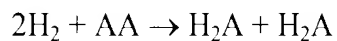
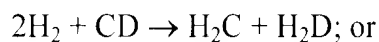
(Amended) In addition, the flow rate of the isotope scrambler return oxygen vapor 116 is extremely low compared with the amount of the ascending vapor within the distillation column to which the oxygen [vapor]is returned. Therefore, it has almost no influence on the distillation efficiency even if it is returned into the distillation column as it is at normal temperature.

On page 33, please amend the paragraph beginning on line 22, as follows.

(Amended) When the whole apparatus of the isotope scrambler 47 is considered, since there are only the conduit 49 for the purified isotope scrambler return vapor 116, and the outlet for the super high purity oxygen which is supplied separately for the purpose of regeneration in the adsorber 96, the other gases[vapors] circulate within the apparatus and there is almost no loss of gas[vapor]. Consequently, the amount of the hydrogen [vapor]and argon [vapor]which are supplied by the supply line for hydrogen [vapor]and argon [vapor]is extremely small.

On pages 33-34, please amend the paragraph beginning on line 28, as follows.

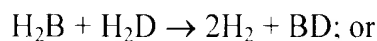
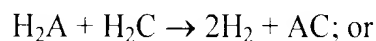
(Amended) Within the catalytic column 88 of the isotope scrambler 47, oxygen and hydrogen are reacted due to the oxidation reaction catalyst and water is produced. For example, if A, B, C, and D are, respectively, any one of the isotope atoms ^{16}O , ^{17}O , and ^{18}O , the atoms which make up the oxygen molecules are separated into different molecules of water by means of reactions like the following.[:]

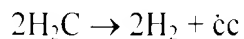


The relative abundance of each of the heavy oxygen isotopes within the reaction product (the water molecules) obtained thereby is determined by the relative abundance of each of the isotopes within the introduction oxygen[output vapor] 115.

On page 34, please amend the paragraph beginning on line 10, as follows.

(Amended) In addition, within the electrolysis apparatus 89 of the isotope scrambler, the water molecules are broken down into the oxygen molecules and to hydrogen molecules by means of electrolysis. In the same way, if A, B, C, and D are, respectively, any one of the isotope atoms ^{16}O , ^{17}O , and ^{18}O , the water molecules are broken down into oxygen molecules and hydrogen molecules by means of reactions like the following.[:]





The combination of the oxygen atoms which make up the oxygen molecules obtained here is[are] determined randomly by the relative abundance of the oxygen isotopes present in the water molecules.

On page 35, please amend the paragraph beginning on line 8, as follows.

(Amended) Accordingly, the abundance ratio of each isotope in the reactant material (i.e., oxygen isotope molecule) obtained by means of isotope exchange by means of the isotope scrambler 47 is determined according to the abundance ratio of each isotope in the introduction oxygen[output vapor] 115.

On page 35, please amend the paragraph beginning on line 11, as follows.

(Amended) In other words, for example, the components of $^{16}\text{O}^{16}\text{O}$, $^{16}\text{O}^{17}\text{O}$, and $^{16}\text{O}^{18}\text{O}$ are present in the introduction oxygen[output vapor] 115. If the respective molar ratios of these components is Y_{11} , Y_{12} , and Y_{13} , the combination of oxygen atoms which make up the oxygen molecules changes randomly based on the abundance probability of each of the isotopes by means of the isotope scrambler 47 using either of the above[-]described methods. the concentration of each component after the isotope scrambling is as follows.[:]

$$^{16}\text{O}^{16}\text{O}:(Y_{11} + Y_{12}/2 + Y_{13}/2)^2 \quad \dots(\text{i})$$

$$^{16}\text{O}^{17}\text{O}:(Y_{11} + Y_{12}/2 + Y_{13}/2)Y_{12} \quad \dots(\text{ii})$$

$$^{16}\text{O}^{18}\text{O}: (Y_{11} + Y_{12}/2 + Y_{13}/2) Y_{13} \quad \dots(\text{iii})$$

$$^{17}\text{O}^{17}\text{O}: Y_{12}^2/4 \quad \dots(\text{iv})$$

$$^{17}\text{O}^{18}\text{O}: Y_{12} Y_{13}/2 \quad \dots(\text{v})$$

$$^{18}\text{O}^{18}\text{O}: Y_{13}^2/4 \quad \dots(\text{vi})$$

On page 35, please amend the paragraph beginning on line 27, as follows.

(Amended) The reaction product in which the concentration of heavy oxygen molecules has been increased using the isotope scrambler 47 is supplied to column $A_i[i]$ through conduit 49 as isotope scrambler return oxygen[vapor] 116, and thereafter passes through columns $A_i[i]$ to $A_n[n]$.

On page 36, please amend the paragraph beginning on line 1, as follows.

(Amended) The heavy oxygen molecules have a higher boiling point and are easily enriched.[,] [t]Therefore, the concentration of heavy oxygen isotopes is further increased in this process.

On page 36, please amend the paragraph beginning on line 6, as follows.

(Amended) In addition, the heavy oxygen isotope enriched material obtained by means of columns $A_i[1]$ to $A_h[h]$ is supplied to the isotope scrambler 47, the concentration of

heavy oxygen molecules within the enriched material is increased by means of isotope scrambling which occurs in the isotope scrambler 47, the enriched material is then supplied to the columns $A_i[i]$ to $A_n[n]$, and the concentration of heavy oxygen isotopes is further increased. Therefore, it is possible to obtain a product containing a high concentration of heavy oxygen isotopes.

On page 36, please amend the paragraph beginning on line 12, as follows.

(Amended) In addition, the apparatus shown in the figure has a structure in which the conduit 49 on the outlet side of the isotope scrambler 47 is connected to the $i^{th}[th]$ column $A_i[i]$. In this way, the column to which the conduit 49 on the outlet side of the isotope scrambler 47 is connected is preferably further down stream (to the later stage side) from the $h^{th}[th]$ column $A_h[h]$ to which the conduit 48 on the inlet side of the isotope scrambler 47 is connected. However, the present invention is not limited to this, and the column to which the outlet side conduit 49 is connected may be the column to which the inlet side conduit 48 is connected or may be upstream (to the earlier stage side) from that column.

On pages 36-37, please amend the paragraph beginning on line 20, as follows.

(Amended) In other words, the isotope scrambler 47 can be inserted at any part of "the apparatus constructed from a plurality of distillation columns" which is constructed in a cascade. For example, it is possible to draw off vapor from any place of the $k^{th}[th]$ column

(including the distillation column, the condenser, the reboiler, the piping, etc.) process it in the isotope scrambler, and return it to any place of the j^{th} column (including the distillation column, the condenser, the reboiler, the piping, etc.). Here, the size relationship between k and j is unrestricted, and it is also possible for the k and j to be equal. However, in order for the isotope scrambling to be conducted efficiently, the place at which the vapor is extracted from the apparatus is preferable a position at which $^{16}\text{O}^{18}\text{O}$ is most enriched. In addition, it is preferable for the position to which the oxygen[vapor] is returned to the distillation column after isotope scrambling to be at a later stage (location at which the $^{18}\text{O}^{18}\text{O}$ is more enriched) than the place of extraction, because the concentration of $^{18}\text{O}^{18}\text{O}$ in the oxygen[vapor] after the isotope scrambling is greater than the concentration of the isotope $^{18}\text{O}^{18}\text{O}$ at the position of extraction.

On page 37, please amend the paragraph beginning on line 4, as follows.

(Amended) In addition, in the present invention, it is possible to obtain heavy oxygen water by means of supplying oxygen extracted from the above_[-]mentioned final column $A_n[n]$ to the above_[-]mentioned hydrogenation reaction apparatus 300, and converting it to water.

On page 40, please amend the paragraph beginning on line 7, as follows.

(Amended) In Conventional Example 1, almost all of the sections of the piping, which are[is] the liquid supply conduits between the first column and the second

column and between the second column and the third column, are full of liquid. In contrast, in Example 1, of the piping (return conduits 14 and 15) which are the liquid supply conduits from the second column to the first column and from the third column to the second column, only a few sections which correspond to the liquid head portion corresponding to the pressure difference for the bottom of the column and the top of the column between the first column and the second column, and between the second column and the third column are full of liquid.

On page 41, please amend the paragraph beginning on line 10, as follows.

(Amended) (2) There is no need to perform maintenance on liquid pumps, or to switch back[]up liquid pumps associated therewith, and therefore it is possible for stable and continuous operation.

On page 41, please amend the paragraph beginning on line 16, as follows.

(Amended) On the other hand, the amount of heat exchange for the condensers and the reboilers is greater in the present embodiment than in the conventional example and this is disadvantageous from[form] the point of view of operation cost. This is because in the present invention the liquid which was the feed from the first column to the second column and from the second column to the third column in conventional apparatuses is vaporized once in the reboiler, and thereafter liquefied in the condenser. In addition, the

vapor which was returned from the second column to the first column and from the third column to the second column in conventional apparatuses is liquefied once in the condenser and thereafter vaporized in the reboiler in the present invention.

On page 42, please amend the paragraph beginning on line 1, as follows.

(Amended) Example 2

Table 4 shows the results of a simulation for a situation of producing heavy oxygen water using water as a starting material and using the apparatus shown in Figure 1. The specifications for the apparatus are shown in Table 3.

On page 46, please amend the paragraph beginning on line 5, as follows.

(Amended) It was assumed that the number (n) of distillation columns was 10 and the isotope scrambler 47 was arranged between the sixth column and the seventh column. The specifications of the apparatus are shown in Table 8. In the table, the isotope scrambler supply vapor indicates the introduction oxygen[output vapor] 115 which is supplied to the isotope scrambler 47 through the conduit 48, and the isotope scrambler return vapor indicates the isotope scrambler return oxygen[vapor] 116 which is drawn off from the isotope scrambler 47 and supplied to column $A_i[i]$ through conduit 49.

On page 51, please amend the paragraph beginning on line 7, as follows.

(Amended) When ^{17}O is not needed and only a product vapor which is enriched in $^{16}\text{O}^{18}\text{O}$ is required, a distillation column having a total height shorter than the present example can be used. In that case, the composition for the peak of $^{16}\text{O}^{17}\text{O}$ is approximately 1%. More specifically, if the composition distribution of $^{16}\text{O}^{18}\text{O}$ of Figure 17 is considered, from the first column to the vicinity of the sixth column, the slope of the composition is small[,] and, in order to enrich $^{16}\text{O}^{17}\text{O}$ to a high concentration, it is necessary to increase the total height beyond that necessary for $^{16}\text{O}^{18}\text{O}$ in the present example. In other words, if the total packing height is increased, it is possible to enrich the intermediate components using the apparatus of the present invention.